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Alexander (Alex) Dalgarno greatly advanced the quantitative study of fundamental atomic and molecular processes, contributed significantly to atmospheric science and 'established molecular astrophysics as a unified intellectual field of great scientific endeavour, impact and achievement' (Flannery 2010). Alex developed and applied techniques that simplify calculations and lead to reliable solutions, enabling him to make landmark contributions to the knowledge of collisionally induced charge transfer, rotational and vibrational excitation of molecules, spin exchange and ultracold chemistry. His wide-ranging curiosity, disciplined steps to broaden his programme and ability to identify dominant physical processes and calculate their rates led to his many important contributions to atmospheric science. For example, Alex greatly expanded the knowledge of terrestrial airglow features, photoabsorption and collisional processes in the terrestrial ozone layer and deposition by energetic electrons in the atmospheres of other planets. In molecular astrophysics, he applied that same systematic approach to studies of a range of environments from the early Universe to present-day UV-irradiated interstellar clouds, shocks and supernova ejecta. Alex made availability to students a priority and encouraged them to pursue problems that they devised, despite his seemingly inexhaustible supply of suitable projects. His community service included his 29-year long editorship of *Astrophysical Journal Letters*, starting in 1973, and the founding of the Institute of Theoretical Atomic and Molecular Physics at the Harvard-Smithsonian Center for Astrophysics, in 1988, owing to his concerns for the health of the fields of theoretical atomic

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and molecular physics and fundamental quantum mechanics, which the Institute did much to reinvigorate.

Early life

Alex Dalgarno* and his twin sister were born in London. He was the youngest of five children, one of whom died young before Alex knew him. His parents, William and Margaret (née Murray), were born and married in Aberdeen, and Alex's grandfathers were a blacksmith and a mill owner, respectively. Alex's father worked as an insurance executive and had been transferred in about 1920 to London where the family lived initially in Wood Green. Alexander Low, a great uncle and professor of anatomy at the University of Aberdeen, declared that 'the male will be brilliant' after performing a phrenological examination of the twin babies (Bates & Victor 1989).

Alex's love of reading led others to describe him as 'The Professor' from an early age. He used his bedroom as a study, where his mother would sometimes serve him dinner. When she looked in on him later, she might find him asleep surrounded by open books.

The family moved to Winchmore Hill, where Alex at age nine passed the 11 plus examination at the local primary school, but was, in his words, 'doomed to repeat each year' (111)† until he became 11. As war approached in 1939 the family was on holiday in Aberdeen, where they remained. For one term Alex attended Robert Gordon's College. However, the family were not satisfied with the living arrangements and returned to London before Christmas.

From January 1940 to July 1945 Alex attended Southgate Grammar School. Throughout the 1940 to 1941 blitz and the sporadic air raids afterwards children sometimes had to use the air raid shelters built near the playing field, but Alex felt that he received a good education and had some excellent teachers. He found the period 'an interesting time, occasionally frightening', but considered the subsequent V1 and V2 attacks of the 1944 mini-blitz 'more threatening' (111). During the mini-blitz he and other boys served on a team responsible for finding incendiary bombs and calling the supervising teacher to extinguish them with sandbags. The boys found such a bomb only once and made the teacher unhappy by dealing with it without him.

In addition to enjoying mathematical puzzles, Alex played football and cricket for the school and local teams and was invited to try out for the Tottenham Hotspur football club. He also played tennis and squash, which he continued into his eighties. In addition, he joined the Air Training Corps.

University College London

In 1945 Alex completed his examinations. Though his father was not enthusiastic about Alex attending university, he applied to study mathematics at University College London (UCL).

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* The sources for much of the biographical material in this memoir, especially that concerning Alex's early life, are his autobiographical essay in *Annual Reviews of Astronomy and Astrophysics* (111) and an article by Sir David Bates and George Victor (Bates & Victor 1989).

† Numbers in this form refer to the bibliography at the end of the text.

After one semester he discovered that he would have to work harder than he had expected and began to work with a level of dedication that subsequently never diminished.

Alex was surprised to pass the mathematics BSc examinations in June 1947 with first class honours. To complete the degree requirements, he attended for another year during which he was on the Advanced Studies programme, became involved in student affairs and played for the UCL football club, of which he was the treasurer. As this period was ending, Alex was uncertain about his next step. Then, during an encounter with Harrie (later Sir Harrie) Massie FRS 1940 in a physics department corridor, Alex was offered a fellowship to support study for a PhD in atomic physics. Alex felt that he knew little about the subject but 'found the prospect of doing research and maybe finding something new very appealing' (111).

Massie suggested that Alex investigate collisions of metastable helium atoms in helium gas under the supervision of Richard Buckingham. Alex decided to perform direct many-electron calculations to obtain the required interaction potentials rather than use empirical potentials. He did the very lengthy calculations with a mechanical calculator. The evaluation of the cross sections showed the long-range interactions to be important (1). Such interactions remained of long-term interest to Alex.

QUEEN'S UNIVERSITY BELFAST

In 1951, roughly as Alex completed the PhD requirements, David (later Sir David) Bates (FRS 1955), a reader at UCL, became head of the Department of Applied Mathematics at Queen's University Belfast (QUB). Responding to Bates's request for advice about appointing an assistant lecturer at QUB, Massey stated that 'Alex was the best research student in an exceptionally good year' (Bates & Victor 1989). Alex started in the temporary post in 1951 and became a permanent lecturer in 1952.

Shortly after arriving in Belfast, Alex and Bates performed the first correct Born treatment of resonant charge transfer in collisions between H(1s) and H^+ (2), a process that Oppenheimer (1928) and Brinkmann & Kramers (1930) had investigated. As mentioned below, over decades Alex developed techniques to calculate charge transfer rates and applied those rates in studies of multiple astrophysical and planetary atmospheric phenomena.

Alex and Bates also investigated the production of electronically excited states of H in collisions between H(1s) and H^+ (5). This work was relevant to the interpretation of observations of proton aurorae, and Bates's suggestion that they perform it was considered by Alex to have affected his future research significantly (111). It demonstrated to him the critical role of atomic and molecular processes in aeronomy and led him to study, with Bates's help, the origins of the nightglow and dayglow. They developed theoretical arguments that enabled them to establish the altitudes of the main features in the airglow (4) and advance the understanding of the dayglow (7). Alex noted that Daniel Barbier said that because of these two papers 'without any change in the observational data the inferred altitudes fell by several hundred kilometres' (111). Alex 'very much enjoyed the combination of observations and experiments interpreted by finding the processes responsible for the phenomenon and calculating or measuring the rates of the processes' (111).

Alex also collaborated with Brian Bransden, then a QUB physics lecturer. Their work included the development of a time-independent variational approach to the calculation of the energies and lifetimes of autoionization states (3) and applications of variational methods to scattering by ions (6). In their final joint paper, which was co-authored with Mike Seaton (FRS 1967) of UCL, they showed that the long-range polarization interaction must be considered in the treatment of electron scattering by neutral hydrogen (14).

In 1954 Alex spent the summer at MIT. They had an electronic computer, which he used to re-calculate the integrals that he had evaluated to obtain the potentials employed in scattering studies that he performed as a postgraduate student. Decades later he wrote, ‘I still remember the joy I felt as those integrals that had taken me hours to calculate streamed out at the rate of one per second. I returned to Belfast determined never to use a mechanical calculator again’ (111). He convinced Bates to seek funding for an electronic computer and ‘played an important part in negotiations with the U.S. Office of Naval Research for a contract that included providing the university with its first computer’ (Bates & Victor 1989). The computer arrived in 1961 and was one of the first two digital computers in Northern Ireland (Jamieson 2010). Alex served as director of the Computing Laboratory until 1965.

While at MIT he calculated the interaction potentials of H and H^-. After returning to Belfast Alex used these potentials with Coulter McDowell, a student, to investigate the mobility of H^- in an atomic hydrogen gas and calculate the cross sections for charge transfer (11). Alex’s first involvement with astrophysics resulted from his interest in H^-, a major source of opacity in the atmospheres of solar-like stars. At a meeting he learned that others believed that the abundance of H^- in the Sun is limited primarily by removal due to electron impact, but Alex realized that associative detachment in reactions of H^- with H is more rapid. He estimated, but did not publish, a value for the rate coefficient. However, in a paper on solar H^-, Pagel (1959) quoted the value and credited Alex with bringing attention to the significance of the process, which is dominant. Subsequently, McDowell (1961) investigated the rate at which the process forms H_2 in the interstellar medium, and Jim Peebles (FRS 1982) and Robert Dicke established its key role in producing pre-galactic H_2, which is important for cooling during structure formation (Peebles & Dicke 1968).

Alex also worked with Alan Stewart, another QUB faculty member. Michael Jamieson, one of Alex’s former postgraduate students, has written, ‘Many of the applications of perturbation theory to investigations in atomic and molecular physics were developed by Alex Dalgarno. The formalism of the perturbation method was published at the start of a very fruitful period by Dalgarno and Stewart’ (Jamieson 2010). In Alex and Stewart’s first joint paper, they re-examined conventional perturbation theory and suggested a combination of variational and perturbation techniques for calculating small disturbances of a stationary system (12). They subsequently published a series of papers concerning applications to phenomena including the Lamb shift in helium, long-range ion–atom interactions, single-photon double ionization, and properties of the helium isoelectronic sequence.

Alex also worked with John Lewis, a student, on perturbation theory. As mentioned above, Alex had already found in his PhD research that the rates of many low-energy heavy particle collisional processes depend on the natures of the long-range interactions. With Lewis, Alex worked to invent methods to obtain precise expressions for the long-range interactions between pairs of species. Leonard Schiff devoted a section in his well-known quantum mechanics textbook to the Dalgarno–Lewis method. In standard perturbation theory the second-order correction to the energy of a state is given by a summation over an infinite number of terms involving matrix elements of the perturbation between that state and each of all of the other states. In the Dalgarno–Lewis approach, the evaluation of that infinite series requires the calculation of only three matrix elements, which involve the unperturbed state, the perturbation potential and the solution to a single differential equation (8). For a long-range interaction, the potential can be written as a sum of terms corresponding to the different multipoles. The Dalgarno–Lewis differential equation for each multipole contribution to the potential can be solved analytically for a perturbed one-electron atom or ion in the ground state (8). Jamieson (2010) summarized the advantages of the Dalgarno–Lewis method over standard perturbation treatments by writing that in their approach 'the excited states and energies are not required explicitly, all of the discrete terms ... are included automatically and the continuum part, which is otherwise difficult to evaluate, is included'.

Subsequently, Alex and Lewis demonstrated the equivalence of their approach and the method based on the variational principle (9). 'This result started the development of variation-perturbation methods' (Jamieson 2010). Later, Alex applied perturbation theory to eigenfunctions and energies obtained with the Hartree–Fock method, in which an approximate potential is initially used to calculate properties of a system with two or more electrons (16). Neal Lane, one of Alex's former postdoctoral assistants, noted that Alex's 'early development ... of a theory which permits the exact evaluation of the series expansion of Hartree–Fock energies in powers of 1/Z provided the foundation for a powerful expansion method for calculating atomic properties' (Lane 1989). Z is the charge of the nucleus of the atom.

With the student Norman Lynn, Alex prescribed how to calculate the coefficient of the long-range van der Waals interaction between two identical atoms from poorly known oscillator strengths, which are proportional to the radiative transition rates, following the imposition of sum rules on the oscillator strengths (13). They applied their method to obtain the van der Waals coefficient for helium, for which they also used the constrained oscillator strengths to calculate the refractive index, the Verdet constant, the diamagnetic susceptibility, the Lamb shift and the average energy loss for fast particle impact. Alex and student Arthur Kingston evaluated the van der Waals coefficients and other constants for other atomic pairs (15, 18). In particular Alex 'gave considerable attention to the quantitative evaluation of the van der Waals coefficient by exploiting the formal connection to the dipole polarizability at imaginary frequencies' (111). With Ron McCarroll, another student, Alex showed that in a diatomic system in which at least one of the atoms is in a state with nonzero orbital electronic angular momentum, the non-adiabatic contribution to the long-range interaction can exceed the van der Waals contribution (10). The non-adiabatic contribution arises from the interaction between the electronic state and the nuclear state, which is assumed to be zero in the Born–Oppenheimer approximation used frequently in calculations of molecular potential curves.

In his highly cited 1962 review concerning atomic polarizabilities and shielding factors (20), Alex emphasized the close relationship between perturbation and variational approaches.

While working on perturbation theory and long-range interactions, Alex made other contributions in atomic, molecular and optical (AMO) physics and also maintained his interest in atmospheric science. In 1955, as requested by Bates, Alex co-organized a conference in Belfast on the airglow and aurorae. The papers concerned 'observations, theory, instrumentation, laboratory studies and quantal calculations' (111). In 1956, he attended a meeting entitled 'The Threshold of Space' organized by the Geophysics Research Directorate of the US Air Force Cambridge Research Center (111). He wrote that 'there was a session on "Rocket Probing of the Upper Atmosphere", in which local measurements were reported and a new era in aeronomy began' (111). He also noted that the results obtained in 1956 and 1958 with rocket-borne mass spectrometers 'established the importance of ion-molecule chemistry in determining the ion composition' in the ionospheric F region and that 'these early rocket flights heralded the coming of the Space Age' (111). At the meeting he met Fred Marmo and Murray Zelikoff, who invited Alex to visit the Air Force Cambridge Research Center to consider possible upper atmospheric experiments with them. He visited for several summers.

The Center provided support for Alex's work with Arnold Arthurs, a QUB student, to develop the S-matrix theory of collisionally induced excitation of molecular rotational levels (17). The work of Blatt & Biedenharn (1952) influenced them and they used tensor algebraic results obtained by Racah (1942). Astrophysicists, atmospheric scientists, AMO physicists and physical chemists have applied the Arthurs and Dalgarno approach in many investigations, and their 1960 paper is Alex's most cited publication.

In the early 1960s, Alex increasingly focused on atmospheric science. He reviewed data for collisions of electrons with atmospheric gases, calculated key photoabsorption cross sections and studied rates of electron-induced rotational excitation of atmospheric molecules (McElroy 1989). With Michael McElroy and Roy Moffett, Alex solved the equations governing the absorption of solar radiation in the atmosphere and obtained rates for photoionization and photodissociation (22). Using a model of the ion chemistry, they calculated the ionization distribution. They also identified the processes governing the thermal balance and showed that in the ionospheric F region the electron temperature exceeds the ion temperature. According to McElroy (1989), this work 'provided the stimulus for a series of follow-up studies, involving both improvements in data for the cross sections of a number of the key processes, in addition to applications of increasing breadth and complexity'. With Jim Walker, Alex considered the excitation of the red line of atomic oxygen in the dayglow (24) and auroral excitation of atomic oxygen forbidden lines (28). He also inferred the energy content of aurorae from the observed intensities of the 391.4 nm band of N_2^+ (25).

From 1962 to 1963 Alex spent a sabbatical year at the Geophysical Corporation of America, which former US Air Force scientists had established, and began interacting with Harvard astronomy faculty, which stimulated him to broaden his research to include astrophysics (111). He had already calculated cross sections for spin exchange in H–H collisions, which govern the spin temperature for the levels involved in 21 cm line emission (19). However, Alex's first research in molecular astrophysics, the discipline of which he is the recognized father (Hartquist & Williams 1998), was his demonstration with the student David Williams that the contribution of Raman scattering by H_2 to the interstellar far ultraviolet (FUV) background is insignificant (21). Alex's subsequent work on processes occurring in H_2 provided key data for the development of models that facilitate the use of H_2 emission and absorption features to diagnose the physical properties of a variety astronomical regions. For example, Alex worked with Arthur Allison to develop numerical methods and computer code to solve the close-coupling equations arising in the Arthurs and Dalgarno treatment (17) of collisionally induced molecular rotational excitation, and made an application to H_2 collisions with H, He and H_2 (27). The quantum data produced by Alex and Allison were used by astrophysicists in four different decades. Alex's earliest considerations of interstellar cooling concerned the contribution of the excitation of low-lying levels of C^+ and Si^+ in spin-flip collisions with atomic hydrogen (23).

Though Alex's activity in atmospheric science and astrophysics increased, he continued to work on AMO physics. For example, he remained interested in long-range interactions. One of several approaches that he used to study them was the coupled time-dependent Hartree–Fock (TDHF) method that he and George Victor developed (26). Though the coupled TDHF approximation including external perturbations had been examined previously, the associated equations did 'not lend themselves to ready solution ... and an alternative formulation of the coupled approximation' was required (26). Alex's development of a practical coupled TDHF method was just one of his important contributions on approximation techniques that enable the calculation of the properties of many-electron systems, and he applied the method to investigate multiple systems.

Harvard and the Smithsonian

In the mid 1960s Alex felt that his 'knowledge of astronomy was limited' (111) and that he would benefit from being somewhere where astrophysics was a major focus. Consequently, he responded positively when Leo Goldberg, the director of Harvard College Observatory, approached him about a professorship in the Harvard Department of Astronomy. In July 1967 Alex assumed the Harvard post and a staff position at the Smithsonian Astrophysical Observatory in Cambridge, Massachusetts.

In the early 1970s, a student considering postgraduate study in astronomy at Harvard received a list of the interests of the Harvard and Smithsonian astrophysicists. Most provided numerous bullet points indicating specific, sometimes closely related, problems. Alex's concise entry contained three items—astrophysics; atmospheric science; atomic and molecular physics—which accurately reflected his broad activity in each of those areas during his entire period in Cambridge, Massachusetts.

Alex recognized the importance of a thorough knowledge of atomic and molecular processes for astrophysics, and in the late 1960s much of his work in astrophysics centred on the provision of such knowledge. Rocket-borne instruments were yielding a wealth of solar spectral data, and the dependence of key solar spectral features on the two-photon decay rates of the $2\ {}^1S$ and $2\ {}^3S$ states of helium-like systems from He I through Ne IX motivated Alex to calculate those rates (32). Rocket-borne and orbiting detectors capable of providing FUV and X-ray spectra of astronomical sources were under development, and soon the detection of FUV absorption features of interstellar H_2 against the nearest high-mass stars would occur. Even before such a detection was achieved, Alex's considerable foresight led him to calculate data for H_2 that would enable the quantitative interpretation of the astronomical H_2 FUV features. The data would also be important for exploiting interstellar H_2 infrared line emission, which was first observed with ground-based facilities in the mid 1970s, to diagnose astronomical sources. The H_2 data important for astrophysics calculated by Alex and collaborators included the oscillator strengths of the Lyman system (30), which are important for the photodissociation and optical pumping of ground electronic state rovibrational levels of interstellar H_2. They also included the rovibrational quadrupole matrix elements for the ground electronic state (31) and the rates at which the $B\ {}^1\Sigma_u{}^+$ levels spontaneously dissociate via bound–free radiative transitions to the vibrational continuum of the $X\ {}^1\Sigma_g{}^+$ state (37). Additionally, Alex identified proton exchange collisions as the dominant source of coupling between *ortho* and *para* levels of H_2 and similar collisions between D^+ and H_2 as key for deuterium fractionation (40).

He began to consider how interstellar molecules form. In the late 1930s CH, CN and CH^+ became the first such molecules to be detected. Distinguished scientists, including Bates and Lyman Spitzer (ForMemRS 1990), had tried to explain their abundances. Bates & Spitzer

(1951) suggested that the radiative association of C^+ with H initiates the reaction sequence forming CH in diffuse clouds. However, the process proved to be insufficient, leading Alex, with John Black, to suggest correctly that the radiative association of C^+ with H_2 initiates the sequence (39). Over the next several years Alex made numerous significant contributions to the development of the gas-phase networks used to interpret observations of interstellar molecules.

He was also interested in the global properties of interstellar matter. Field *et al.* (1969) introduced an isobaric, steady-state, hydrostatic, two-phase model of the interstellar medium that accounted for the existence of pressure-bound clouds embedded in a roughly 10^4 K intercloud medium with properties consistent with existing 21 cm observations. Alex, Chris Bottcher, Mike Jura and Dick McCray noted that the timescale of variations in the ultraviolet background due to supernovae is shorter than the timescale to establish pressure equilibrium on intercloud scales, and they developed a time-dependent model of the interstellar medium (33). Subsequently Cox & Smith (1974) and McKee & Ostriker (1977) constructed models including the time-dependent injection of mechanical energy by supernovae. Alex and McCray produced an influential review of the interstellar heating, ionization and cooling processes governing the temperatures of the different interstellar phases (36).

While becoming an established astrophysicist, Alex remained a leading atmospheric scientist. He began to study extraterrestrial atmospheres and applied his work on H_2 radiative processes to consider H_2 photodissociation on Venus (29). He considered the implications of Mariner 6 and 7 observations of Martian airglow. For example, he concluded that the production of electronically excited $CO_2{}^+$ during the photoionization of CO_2 is the dominant mechanism causing some of the ultraviolet features (34). His most important efforts concerning atmospheres then were connected with his leading role in the development of the Atmosphere Explorer Program (41, 42). In a special issue of *Planetary and Space Science* dedicated to Alex, when describing Alex's collaborations on the interpretation of data obtained with Explorer 17 and on the Atmosphere Explorer Program planning, Walker (1988) wrote:

> These collaborations were so fruitful that he became a Co-Investigator for the Langmuir Probe Experiment and a theoretical member of the science team for the Atmosphere Explorer Program. This was the first time that theorists had been included from the outset as full members of the investigator team of a NASA satellite mission. Alex Dalgarno served with distinction for a number of years as Chairman of the Atmosphere Explorer Program's Aeronomy team.

The Atmosphere Explorer missions provided simultaneous measurements of the atmospheric composition, the electron energy distribution and the solar ultraviolet flux and made possible the first check of the reliability of ionospheric chemistry models (McElroy 1989).

Alex continued to conduct research in AMO physics with no immediate astrophysical applications. During the early 1970s he and co-workers developed model potential techniques with which a multi-electron system is treated as an electron or a set of valence electrons in an effective potential created by the nucleus or nuclei and the other electrons. He and Tom Caves adopted a semi-empirical model potential including core-polarization terms and, by varying parameters and solving the one-electron Schrödinger equation for the model potential, reproduced measured atomic energies. They used the resultant eigenfunctions to calculate dynamical polarizabilities, oscillator strengths, photoionization cross sections and radiative recombination rates (35). With Bottcher, Alex introduced a model potential approach based more closely on first principles and less dependent on *ad hoc* assumptions. They addressed diatomic molecules as well as atoms (43).

Remarkably, Alex carried a heavy administrative load while conducting exceptional research in multiple broad areas. He served as chairman of the Department of Astronomy from 1971 to 1976, acting director of Harvard College Observatory from 1971 to 1973 and associate director of the Center for Astrophysics from 1973 to 1980 (Bates & Victor 1989). In 1971 he became the editor of *Astrophysical Journal Letters*, which he remained for 29 years. He wrote, ‘It seemed a very good way to learn astrophysics and so it turned out, though I learned still more about people’ (111).

In 1973 the publication of the first spectroscopic results for interstellar H_2 obtained with the Copernicus satellite (Spitzer *et al.* 1973) and the launch of Atmosphere Explorer C (42) resulted in the emphasis of a good fraction of Alex’s work in astrophysics and aeronomy shifting from being preparatory to interpretative.

In work with John Black, on the abundance of deuterium in a diffuse molecular cloud towards the high-mass star ζ Oph, Alex made his first use of Copernicus ultraviolet data (38). The cosmic elemental abundance of deuterium depends sensitively on the properties of the Big Bang. Thus, there was interest in the deuterium abundance as a diagnostic of the nature of the Big Bang. However, deuterium is removed in stars, which might cause regional abundance variations. The interpretation of Copernicus data enabled the study of such variations. Alex and Black interpreted observations of HD, the abundance of which depends on the cosmic-ray-induced ionization rate in a cloud and on the deuterium abundance. They noted that the OH abundance can be used to infer the ionization rate since cosmic-ray-induced ionization initiates OH production. If one knows the properties (e.g. the density) of a cloud and its radiation environment, OH and HD data can be used to infer the deuterium abundance. To study those properties of the ζ Oph and other clouds, Alex and Black calculated the response of the H_2 infrared emission and the rotational population of the lowest vibrational level of the H_2 ground electronic state to ultraviolet radiation (47) and interpreted rotational H_2 level populations obtained with Copernicus (48). Alex, Black and their collaborators used this approach to investigate the variations of the interstellar ultraviolet background, physical properties of diffuse molecular clouds, ionization rate and deuterium abundance between regions.

The ionization rate is key for the thermal balance of the cold and warm phases of the interstellar medium and the initiation of much of the gas-phase chemistry. In dark regions destined to host stellar birth, the ionization is induced predominately by cosmic rays. In them the non-ideal magnetohydrodynamic diffusivities affecting processes including angular momentum transport, formation of structure due to non-linear steepening of waves and gravitationally induced collapse depend on the fractional ionization. Alex often stated that astrophysical molecular processes are important for governing dynamics as well as for their diagnostic utility and intrinsic interest to quantum physicists and chemists. With Michael Oppenheimer, Alex showed that the charge transfer of molecular ions with trace metallic atoms limits dissociative recombination, investigated the role of collisions with grains in ion recombination and derived a frequently used analytic prescription for the fractional ionization (45).

Describing Alex’s work of the mid 1970s, with collaborators including Kate Kirby, Oppenheimer and Victor, on the interpretation of Atmosphere Explorer results, McElroy (1989) wrote: ‘It was an intense, interactive experience, documented in a remarkable series of papers ... Atmosphere Explorer changed the state of upper atmospheric aeronomy in five brief years ... An entire area of research came of age and Alexander Dalgarno played a major role in its transformation.' In one of the later papers of that series, Alex and collaborators presented a description of the processes producing and removing O^+ in the upper atmosphere and a model of the ion chemistry in the quiet ionosphere (49). They predicted altitude profiles of the O^+, $N_2{}^+$, NO^+ and $O_2{}^+$ abundances that are very similar to those observed at altitudes of 140 to 250 km with Atmosphere Explorer C.

In the mid 1970s frequent press coverage of the depletion of stratospheric ozone enhanced public awareness of the importance of the propagation of ultraviolet radiation in the atmosphere. The dominant source of atmospheric opacity in the wavelength range from 175.0 to 205.0 nm is due to discrete lines in the Schumann–Runge band system of O_2, and Alex calculated the associated opacity distribution functions (44), which have been used widely in atmospheric models.

Alex's interest in extraterrestrial atmospheres grew. He noted that the theoretical approach applied in the exploitation of Atmosphere Explorer data could be applied to other planets (111). He, Victor and Tom Cravens studied the energy deposition of electrons, produced by the ionization of H_2 induced by cosmic rays or particles accelerated in the interaction of the solar wind with a planetary magnetosphere (46). This deposition is important for heating and ionization in the atmospheres of the Jovian planets and for interstellar molecular clouds.

In the second half of the 1970s, with Walter Johnson, Chi-Dong Lin and Philip Shorer, Alex developed and applied the relativistic random phase approximation for the study of atomic structure. Relativistic effects are particularly significant in an atom with a nucleus with many protons. One of Alex's key applications of the method was to the radiative decays from $n = 2$ levels of ions in the He isoelectronic sequence (50). Such transitions are observed to diagnose high-temperature laboratory and astrophysical plasmas. Many of the useful features occur at X-ray wavelengths.

Slightly later, with Scott Butler and Tim Heil, Alex returned to the subject of charge transfer. They reported the results of quantal calculations of the rates of charge transfer between multiply charged ions with neutral hydrogen and helium atoms (53). They also considered the spectral lines emitted during the reactions. Charge transfer also affects the plasma emissivity by altering ionic abundances. In several studies, Alex, Butler and Heil employed a general procedure, developed by Alex and Heil (52), applicable to any number of states for transforming from an adiabatic basis set to a diabatic one. If strong coupling between adiabatic states obtains, the advantage of the appropriate diabatic description is that off-diagonal matrix elements are small and transitions can be analysed more easily.

Until the late 1970s, most molecular astrophysics investigations concerned interstellar clouds with temperatures below about 100 K. From the late 1970s until the 1980s Alex focused much of his astrophysical research on warmer, very dynamic molecular regions. Infrared line features of H_2 originating in shocked gas had been detected in a star-forming region in Orion (Gautier *et al.* 1976) in which some molecular species produce line emission features having pronounced high-velocity wings an order of magnitude wider than the wings of features emitted by many species elsewhere (Zuckerman & Palmer 1975). The broad wings arise in the shocked gas, in which the relative abundances of some molecular species differ significantly from those in more quiescent interstellar gas. The potential importance of shocks driven by outflows of young stars in inducing further star formation was recognized.

Alex, Hartquist and Oppenheimer developed a chemical model to account for the particularly high fractional abundances of SiO and some sulfur-bearing molecules in the high-velocity gas (54). Alex and Wayne Roberge calculated the temperature and density dependences of the collisionally induced dissociation of H_2 and elucidated the consequences of that process for the molecular composition in shocked gas (56). Through his identification of key collisional and radiative processes and provision and compilation of the corresponding data, Alex made major contributions, with Bruce Draine and Roberge, to the development of multi-fluid magnetohydrodynamic (MHD) models of shocks in diffuse interstellar clouds and denser star-forming regions (57).

By the mid 1980s, the origin of interstellar CH^+ had already been an outstanding problem for nearly five decades. In 1966 Terry Carroll and Edwin Salpeter (ForMemRS 1993) suggested that CH^+ would have a high abundance in shock-heated gas (Carroll & Salpeter 1966). Alex, David Flower, Hartquist and Guillaume Pineau des Forêts constructed a multi-fluid MHD model of diffuse cloud shocks to explore whether observed interstellar CH^+ might have such an origin (62). The effects of the chemistry on the shock dynamics were included self-consistently. It was concluded that the production in shocks of the observed CH^+ abundances would be challenging to reconcile with other observational constraints. The investigation of alternative dynamical models of interstellar CH^+ production continues.

In parallel with the shock studies, Alex, with Stephen Lepp, addressed the deuteration of molecules in dark molecular regions, including those where stars form (61). In such regions, having temperatures much below 100 K, the abundance ratios of deuterated to protonated isotopologues of molecules, other than molecular hydrogen, are often two or more orders of magnitude greater than the cosmic deuterium to hydrogen ratio. The enhanced relative abundances of deuterated species are due to their zero-point vibrational energies being lower than those of their protonated counterparts (Watson 1974). Alex and Lepp realized that substantial fractions of deuterium are in both atomic D and HD and included both species in their chemical network. The dissociative recombination of deuterated ions with electrons limits the abundance ratios of deuterated and protonated isotopologues, and for several sources Alex and Lepp constrained the fractional ionizations from measured isotopologue abundance ratios.

Alex also examined the consequences of the presence of large molecules, including polycyclic aromatic hydrocarbons, on the ionization and other properties of diffuse interstellar clouds. He, Black, Lepp and van Dishoeck showed that the inferred abundances of elements such as lithium, sodium or potassium depend significantly on the abundances of the large molecules because they are recombination sites (63).

In 1984 Alex led the successful effort to establish the International Astronomical Union astrochemistry working group. The group organizes international conferences, and its founding helped to gain recognition for astrochemistry as a significant component of astronomy (figure 1).

In the late 1970s and the 1980s Alex's contributions to atmospheric sciences included his participation in the interpretation of data obtained during the Voyager fly-bys of Jupiter and with probes to Mars and Venus (McElroy 1989). Alex and Jane Fox constructed a chemical and one-dimensional structural model of the Martian atmosphere at altitudes of 100–240 km (51). They designed the model to reproduce the measured altitude profile of the CO_2 abundance and used it to generate extensive results for airglow emissions from multiple species. They were able to match the measured profiles of ${O_2}^+$, ${CO_2}^+$ and O^+, predict the profiles of

Figure 1. Alex in Udaipur in 1985 with Ewine van Dishoeck and Stephen Lepp near the time of the first International Astronomical Union Symposium on Astrochemistry, which was held in Goa. (Image: Ewine F van Dishoeck.) (Online version in colour.)

Ar^+, N_2^+ and NO^+, and assess the contributions of neutral heating due to the chemistry, photodissociation, quenching and electron-impact-induced dissociation. They also developed an analogous model for the upper atmosphere of Venus (55). McElroy (1989) considers Alex's and Fox's work (58) on the escape of nitrogen, which is indicated by the enhanced abundance of ^{15}N relative to ^{14}N, from Mars to be particularly notable. Insight into the mechanisms producing atoms that move fast enough to escape bears on the understanding of the history of volatiles. Alex and Fox concluded that the primary source of fast atoms is the dissociative recombination of ground state and vibrationally excited N_2^+ and were able to account for the ^{15}N to ^{14}N abundance ratio.

In the late 1970s and the middle 1980s Alex, with numerous collaborators, performed important studies of the photoionization and photodissociation of molecules. Many are relevant to interstellar or atmospheric chemistry. The 'photodissociation studies are particularly interesting from a fundamental perspective' (Lane 1989). Alex collaborated with van Dishoeck on OH photodissocation, which is important in interstellar and cometary chemistry. They investigated the roles of multiple excited electronic states for each of several symmetries and found that transitions to discrete levels that either predissociate or decay radiatively to repulsive electronic states contribute along with transitions to repulsive states (59). They concluded that the mechanism previously thought to dominate OH photodissocation in interstellar clouds is insignificant and that near ionizing interstellar shocks Lyman alpha radiation contributes significantly to OH photodissociation. They, Allison and Marc van Hemert performed the first fully quantum calculations including coupled final electronic states in their study of resonances in the OH photodissociation cross section arising from the interaction between $^2\pi$ electronic states (60).

THE ITAMP ERA AT HARVARD AND THE SMITHSONIAN

Alex became concerned by the absence of theoretical AMO physics in those US physics departments that train most physicists who become permanent faculty members in US universities (111). He considered theoretical AMO physics and fundamental quantum mechanics to be disciplines in grave danger of vanishing. Consequently, Alex, Lloyd Armstrong and Neal Lane organized a gathering of AMO theorists in May 1984 at the annual meeting of the Division of Electron and Atomic Physics. This led to the establishment of the Theoretical Atomic, Molecular and Optical Community (TAMOC), a review by a National Research Council (NRC) committee and a 1987 NRC report containing the recommendation that a theoretical AMO physics institute, which would attract graduate students to the field and promote interactions between members of the international AMO community, be created.

With Kirby's and Victor's help, Alex wrote a proposal to the National Science Foundation for funding for the Institute for Theoretical Atomic and Molecular Physics (ITAMP). The name later included 'Optical', though the institute continued to be known as ITAMP (111). The proposal contained Alex's statement that 'physics is embodied not in its equations, but in the solutions to the equations' (Hartquist 1990), a view also expressed in Alex's autobiographical essay (111).

In November 1988 Alex became the first director of ITAMP. Its long-term members have included Harvard and Smithsonian scientists working at the Center for Astrophysics and professorial staff in the Harvard physics and chemistry departments. In 1993 Alex stepped down as the director but remained an active ITAMP member.

Kirby (2010) has summarized the first 20 years of ITAMP. Key appointments included those of Rick Heller and Mikhail Lukin to Harvard professorships, Jim Babb and Hossein Sadeghpour to permanent Smithsonian positions, and postdoctoral researchers who subsequently assumed teaching and research positions at universities and research establishments in the US and elsewhere. ITAMP has had an extensive visitor programme, which has stimulated and enabled collaborations and leading research. During the first ITAMP decade, Alex was particularly productive in AMO physics.

His interest in charge transfer continued, and in that decade he produced at least a half dozen papers on charge transfer rates with co-authors including Kirby, Lane, Sadeghpour, Vasili Kharchenko, Mineo Kimura, Yan Sun and Bernard Zygelman. Alex also continued to study photoabsorption processes. For example, with Sadeghpour he calculated the ratio of the rates of photoionization processes that eject two electrons and one electron in neutral helium and two-electron ions (71). Ultraviolet and X-ray spectral features of two-electron systems provide diagnostics of many astrophysical and laboratory plasmas. Alex also studied the production of several molecules by radiative association. Some of those studies relevant to ultracold collisions, astrophysics and atmospheres are mentioned below.

With the development of techniques to investigate atomic and molecular interactions in ultracold traps, Alex returned to long-range interactions because of their crucial role in low-energy collisions (Jamieson 2010). To second-order accuracy in perturbation theory, Alex, Mircea Marinescu and Sadeghpour calculated the dispersion coefficients for the dipole–dipole,

dipole–quadrupole, dipole–quadrupole and quadrupole–quadrupole terms for interactions in alkali-metal dimers (72). They presented 'a method for computing dynamic multipole polarizabilities at imaginary frequencies that is exact given the assumption of a model potential for the motion of the valence electron in the presence of a frozen core. The infinite second-order sums are transformed into integrals over the solutions of two coupled inhomogeneous differential equations' (72). Their method is of wide relevance, including to the production of Bose–Einstein condensates (Leggett 2001) and the use of Rydberg atoms 'to implement quantum gates between neutral atom qubits' in quantum information research (Saffman *et al.* 2010). To treat electron–electron correlations accurately in calculations of dispersion coefficients of other systems of interest in ultracold physics, Alex, Zong-Chao Yan, Babb and Gordon Drake adopted a variational approach (75).

While studying long-range interactions, Alex investigated ultracold collisions (Balakrishnan 2010, Côté 2010). According to Roman Krems, Alex's 'influence on the development of the research field of cold atoms and molecules is so immense that solving any significant problem in this research field feels like walking in his footsteps' (Krems 2010). After Alex and Robin Côté studied elastic collisions of two ^{7}Li atoms and then of two Na atoms, they explained behaviour observed in the photoassociation spectrum of ultracold atoms, which Randy Hulet had described to Alex. Consequently, Alex, Côté and Yan Sun developed a procedure for inferring the values of positive scattering lengths from photoassociation spectra arising from ultracold collisions (73). This has utility because, for low-energy elastic scattering, the cross section is 4μ times the scattering length squared. Alex and Côté also theoretically demonstrated how stable ultracold molecules can be created by the irradiation, by a laser at an appropriate frequency, of the excited molecules produced by radiative association (84). With Hulet, Eric Abraham and others, they used the results of two-photon photoassociative spectroscopy to infer values of scattering lengths and found a particularly large value for that of ^{6}Li–^{6}Li collisions, which they concluded indicated the existence of a near-threshold resonance (81). Such a Feshbach resonance occurs when the energy of the collision partners equals the energy of a closed-channel bound state. Now 'Feshbach resonances are the essential tool to control the interactions between atoms in ultracold quantum gases' (Chin *et al.* 2010). With Naduvalath Balakrishnan, Robert Forrey and Kharchenko, Alex introduced a multichannel effective range theory suitable for treating ultralow-energy scattering in the presence of resonances and showed that such scattering can be characterized by a complex scattering length (83). Its imaginary part is related to the inelastic cross section. It also allows the derivation of the energies and lifetimes of the quasi-bound states, and Alex and his co-authors noted that it gives the rate of decay of the corresponding Bose–Einstein condensate. Additionally, they showed that Feshbach resonances occur in ultracold atom–diatomic collisions (88).

Collisionally induced rovibrational de-excitation can cause significant molecular losses from traps used in ultracold experiments (95). Though the results given by Wigner (1948) imply that threshold rate coefficients for inelastic scattering and chemical reactions are finite (83), even in the late 1990s the magnitudes of those rates were unknown (Doyle *et al.* 2004). The first calculations of rate coefficients for vibrational de-excitation due to atom–molecule collisions (Schwenke & Truhlar 1985) were for a system with small coefficients at threshold (Doyle *et al.* 2004). In work with Balakrishnan and Forrey on H_2 vibrational de-excitation in collisions with H (82) and with ^{3}He and ^{4}He (87), Alex initiated a programme on rovibrational de-excitation induced by low-energy collisions. They showed that the vibrational energy transfer can be efficient under ultracold conditions and depends sensitively on the initial vibrational level.

During the first ITAMP decade, Alex continued to make many contributions in astrophysics, including: the provision, with collaborators including Lepp, Roberge, Roland Gredel, Eric Herbst, Dylan Jones, Ionel Simbotin and Lutoslaw Wolniewicz, of data used in the modelling of molecular sources (64, 69, 91); the calculation, with David Neufeld, of the structures and chemistry of shocks that dissociate molecules (65); the investigation, with Amiel Sternberg, of the infrared and chemical responses to ultraviolet radiation of molecular regions with sufficient densities that collisional de-excitation of H_2 vibrational levels occurs at rates comparable to or greater than radiative de-excitation (66, 74); and the study, with Gredel, Lepp and Stefano Tiné, of the infrared and chemical responses to X-rays of molecular regions (77, 85).

Alex showed that molecular astrophysics is a broad topic extending beyond interstellar clouds and star-forming regions. With Lepp and Phil Stancil, Alex re-examined the lithium chemistry in the early Universe (78) because lithium-bearing species had been proposed to influence cosmic microwave background anisotropies. However, they concluded that LiH and LiH^+ affect primary anisotropies insignificantly. The energy of the lowest excited rotational level of HD playing a role in cooling is much less than that of the corresponding rotational level of H_2, which stimulated interest in HD cooling in primordial cloud formation. Consequently, Alex, Lepp and Stancil provided ‘a thorough survey of all potentially important gas-phase reactions involving the primordial elements produced in the Big Bang, with a particular emphasis on deuterium’ (90).

Lepp and Stancil are among those who collaborated with Alex on the chemistry of supernova SN 1987A ejecta. The initial model ejecta CO emission luminosity obtained by Alex, Lepp and McCray was too large (68). Alex, Lepp and Weihong Liu noted that that luminosity is reduced when departure from local thermal equilibrium and optical depth effects are included (70). However, they also had to invoke microscopic mixing between the ejecta layer consisting primarily of helium, and that where CO is formed. Such mixing leads to He^+ removing CO. The Rayleigh–Taylor instability, which occurs in the ejecta, deforms the shape of the surface between two different layers, but the degree of microscopic mixing that results is unknown. Thus, the CO emission provides otherwise unavailable insight into the microscopic dynamics of a supernova explosion.

Like some early Universe chemistry, much of the ejecta chemistry is initiated by radiative association reactions. With various collaborators, including Mengli Du, Kirby and Stancil, Alex calculated the corresponding rates required in both areas (67, 76).

During the first ITAMP decade, Alex maintained his activity in atmospheric science. The interpretation of NO rotational excitation data for the terrestrial thermosphere obtained with the CIRRIS 1A space shuttle mission triggered Alex and Kharchenko to establish a long-term programme to study theoretically the kinetics of the thermalization of fast particles produced by photochemistry. Prior to their work with Jathinus Tharmamel (86), investigations of the thermalization of fast N atoms by collisions with O were based on the Hard Sphere Approximation (HSA) for scattering. Alex, Kharchenko and Tharmamel used quantum scattering theory to calculate cross sections employed to construct the kernel in the Boltzmann equation which they solved to obtain the steady-state distribution function for fast N atoms. Their approach gives a significantly larger density of N atoms at higher energies than obtained for the HSA.

Alex and Liu modelled the photoelectron-impact- and solar-fluorescence-induced H_2 and HD contributions to the ultraviolet Jovian dayglow (79) and the Jovian auroral ultraviolet emission induced by the electron impact excitation of H_2 (80). With Kharchenko, they performed Monte Carlo calculations of the energy, charge and excitation distributions of oxygen ions that precipitate in the Jovian atmosphere (89). They included charge exchange collisions. Using their calculated distributions, they modelled the X-ray and EUV spectra of the oxygen ions due to radiative cascades from highly excited states of the oxygen ions.

During the second ITAMP decade, Alex continued to investigate ultracold collisions. He, Balakrishnan, Forrey, Michael Haggerty and Heller studied quasiresonant energy transfer effects in atom–diatom collisions (92). Quasiresonance refers to the dominance in collisions of simultaneous and related specific changes of vibrational and rotational quantum numbers of the diatom. They showed that, in atom–diatom collisions, quasiresonant transitions occur even as the collision energy approaches zero, but that threshold effects can be important and some quasiresonant channels can close. Extending earlier work on He–H_2 collisions to include rotational as well as vibrational transitions, Alex, Balakrishnan and Forrey further elucidated the role that resonances play in rovibrational de-excitation in ultralow-energy atom–diatom collisions (95). They found that in He–H_2 collisions, shape resonances arising owing to the van der Waals well in the potential significantly influence the quenching at energies less than the well depth. Their work also revealed Feshbach resonances near the thresholds for channels of the $v = 0, j = 1$ and $v = 1, j = 1$ levels, where v and j are the vibrational and rotational quantum numbers, respectively, and that these Feshbach resonances greatly affect the limiting values of the elastic and rotational quenching cross sections for $j = 1$ levels. Like collisionally induced rovibrational de-excitation, collisionally induced spin depolarization can cause molecular losses from magnetic traps used in ultracold experiments, which motivated Alex to study with Krems spin depolarization due to atom–molecule and molecule–molecule collisions in magnetic fields (107). The formalism that they employed differs from that of Arthurs and Dalgarno (17) and uses a fully uncoupled space-fixed basis set representation (Krems 2010). They identified the primary mechanisms causing spin depolarization in collisions of ^1S atoms with $^2\sum$ and $^3\sum$ molecules, $^2\sum$ molecules with $^2\sum$ molecules and $^3\sum$ molecules with $^3\sum$ molecules. Alex and Côté investigated charge transfer in Na^+–Na collisions and showed that even at a few degrees Kelvin charge transfer is rapid and might enable efficient cold ion production (96).

Alex, in work with Balakrishnan on $F + H_2 \rightarrow FH + H$, turned his attention to reactive scattering at ultracold temperatures (100). They discovered that ultracold chemistry occurs. They showed that for the $F + H_2$ system at ultralow energies, the van der Waals interaction and quantum mechanical threshold effects govern the rate coefficient and that the inclusion of the fine-structure splitting of F reduces the threshold rate coefficient by a factor of about 5. Subsequently, Alex, Balakrishnan, Eric Bodo and Franco Gianturco showed that near threshold the reaction of F with D_2 $(v = 0, j = 0)$ is about two orders of magnitude slower than that of F with H_2 $(v = 0, j = 0)$. They attributed the result to the presence of a virtual state close to the threshold for the latter, but not the former, system (104).

Alex, Piotr Froelich, Svante Jonsell, Alejandro Saenz and Zygelman investigated collisions of hydrogen atoms with antihydrogen atoms. Alex was motivated by the anticipation of progress being made in the experimental production and trapping of antihydrogen atoms at low temperatures and the possibility that this could facilitate tests of fundamental physical principles including CPT invariance and the weak equivalence principle (101). They showed that, at low energies, the elastic scattering length of hydrogen–antihydrogen collisions is 18 times larger than that of hydrogen–hydrogen collisions. They found that at temperatures below about 0.05 K cooling of antihydrogen by collisions with hydrogen becomes inefficient owing to the increase with decreasing temperature of the ratio of the inelastic and elastic cross sections. The primary inelastic processes are annihilation and a rearrangement reaction yielding protonium and positronium.

Alex continued to study long-range interactions required in calculations of rate coefficients for ultralow-energy collisions. For example, with Babb and Andrei Derevianko, he used a variety of relativistic methods and high-precision experimental data to obtain van der Waals coefficients for heteronuclear alkali-metal dimers (99). Alex and Xi Chu described a linear response time-dependent functional theory and applied it to the calculation of the dynamical polarizations and van der Waals coefficients of complex atom pairs (105). A functional is a function of another function. For example, a model effective single electron potential is a function of the total electron density, which is a function of time and position. Polarizabilities given by density functional methods are particularly valuable for systems for which more precise calculations would be too computationally demanding (Mitroy *et al.* 2010).

Alex's astrophysical research during the second ITAMP decade included a re-examination, with Liu and Min Yan, of energy deposition in a weakly ionized mixture of H, H_2 and He by electrons produced by X-ray- or cosmic-ray-induced ionization (94) and the refinement and extension (102), with Lepp and Stancil, of their model (90) of early universe chemistry (Figure 2). He also critically reviewed the inferred rates of cosmic-ray-induced ionization in the interstellar medium (108). Alex, Liu and Donald Clayton showed that radioactively induced ionization and dissociation of CO leads to carbon dust growth by carbon radiative association in supernova ejecta even if they are oxygen-rich (93).

Cometary plasmas are cold and might seem unlikely X-ray sources. However, Lisse *et al.* (1996) discovered cometary X-ray emission. The discovery triggered collaborations in which Alex and Kharchenko, with others, investigated the emissions induced by charge transfer between solar wind ions and ambient Solar System plasmas. Their work on cometary emission (97) explored a suggestion made by Cravens (1997) and enabled them to infer the relative abundances of some ions in the solar wind (103). They showed that time-variable 0.5–0.9 keV emission observed towards the dark side of the Moon is due to charge transfer between solar wind ions and H in the geocorona (106). Donald Cox had earlier recognized that such geocoronal emission and the charge transfer of solar wind ions with plasma throughout the heliosphere could contribute significantly to the soft X-ray background (Cox 1998), which had been attributed primarily to a local hot interstellar bubble created by at least one supernova. Consequently, Alex, Kharchenko and collaborators calculated self-consistent distributions in the heliosphere of interstellar hydrogen and helium and solar wind ions for a steady solar wind and from them produced synthetic emission maps and spectra (109). They could reproduce the 0.52–0.75 keV emission lines observed during the XMM-Hubble Deep Field North exposure without any contribution from beyond the heliosphere.

During the second ITAMP decade, Alex maintained some activity in atmospheric science. With Kharchenko, Zygelman and Jeng-Hwa (Sam) Lee, he continued to study the kinetics of the energy transfer of fast particles in the terrestrial atmosphere in work on energy transfer in collisions involving fast oxygen atoms (98). He, Kharchenko, Stancil and David Schultz showed that the spectra of Jovian polar X-ray aurorae are consistent with them arising from the precipitation of energetic oxygen and sulfur ions of magnetospheric origin and which have

Figure 2. Alex, with Stephen Lepp, Ted and Elizabeth Stecher, Ant Jones and Antonella Natta at the July 2002 meeting honouring David Williams held at Cumberland Lodge, Windsor Great Park. (Image: David Williams.) (Online version in colour.)

been accelerated to energies of the order of an MeV per nucleon (110). These ions induce atmospheric ionization and charge transfer with atmospheric species.

ALEX'S FINAL ITAMP YEARS

ITAMP began its third decade in November 2008, two months before Alex turned 81. From 2009 to 2014 he authored or co-authored 48 publications. The most significant in atmospheric science and astrophysics included those containing reports of the results of refined models of the Jovian polar X-ray aurorae (112) and charge-transfer-induced heliospheric X-ray emission (113). In an investigation of ultracold NH collisions with N, he and his colleagues concluded that it may be possible to cool NH, and other highly polar molecules in their ground $^2\sum$ states, in useful quantities to milli-kelvin temperatures (115). Such cooling enables potential applications of trapped ultracold species. Alex, Tommaso Calarco, Krems, Igor Lesanovsky, Jörg Schmiedmayer and Timur Tscherbul wrote a key paper on ultracold collisions in external fields. It concerns inducing Feshbach resonances with radio frequency magnetic field fluctuations, which can be tuned to control a resonance. They concluded that shifting Feshbach resonances with such fluctuations might facilitate 'the preparation of exotic many-body states and novel quantum phases' (114).

Jérôme Loreau, Stéphane Vranckx, Michèle Desouter-Lecomte and Nathalie Vaeck co-authored Alex's final refereed paper. It contains results for the radiative association and photodissociation of HeH^+ in the metastable 2 ^3S state (116). HeH^+ interested Alex for over 30 years because it was the first molecular ion to form in the early Universe and was potentially detectable in some planetary nebulae or other galactic sources (116). Güsten *et al.* (2019) cited several of Alex's publications, including his final refereed paper, when they reported the first detection of astronomical HeH^+ emission after observing the planetary nebula NGC 7027.

In 2014, Alex's final publication, which he co-authored with Loreau and Sadeghpour, appeared in the proceedings of an International Conference on Photonic, Electronic and Atomic Collisions. It contains results, relevant for attempts to cool Ag in a magneto-optical trap, for the potential curves for Ag(5s) and Ag(5p) interacting with noble gases (117).

Alex began his career working in atomic and molecular physics and, although he made immense contributions to atmospheric science and astrophysics, he ended his career with a contribution to the field in which he started.

Appreciation

The quantity, range and quality of Alex's contributions create a challenge for someone summarizing them, and Alex obtained numerous striking results on problems other than those mentioned in this memoir. Throughout most of his career, Alex maintained high levels of activity simultaneously in AMO physics, atmospheric science and astrophysics. Of his approximately 850 publications, about 56, 15 and 29% concerned each of those areas, respectively. He obtained major awards from professional societies for astronomy, chemistry, geophysics and physics. One could devote a memoir to his work in each of three broad disciplines. In each he often simultaneously pursued multiple themes. Some of those themes, including long-range interactions and charge transfer, received his attention during multiple periods of his career.

The manner in which Alex developed a theme or discipline was remarkable. His deep knowledge of basic molecular processes was always at the heart of his achievement of new insights. His work in molecular astrophysics provides one example. In the 1960s he foresaw a need for rates and rate coefficients well before the relevant observations were obtained. He calculated those data, which sometimes required that he develop the relevant quantum theory and introduce new methods. He then constructed models, which incorporated those data and which he subsequently employed to interpret observations to diagnose the properties of astrophysical sources and identify the processes governing them. He showed vision, which he maintained over the extended period required to conduct the several phases of work necessary to bring his project to fruition. His efforts in atmospheric science displayed characteristics similar to those of his work in molecular astrophysics. Alex's interest in long-range interactions, which began with his first research, led to his fundamental contributions to perturbation theory and decades later to his deep and comprehensive programme on ultracold collisions and ultracold chemistry. His work on those related themes demonstrates how he would establish a rich evolving enterprise through a thoughtful, systematic progression. The full appreciation of that point requires a consideration of multiple papers and their relationships to one another.

Alex made his availability to students a priority. He was the ideal supervisor. He could identify problems that were suitable no matter what the student's ability was and whether the student was an undergraduate or an advanced postgraduate. He was patient and provided insightful guidance when a student encountered a barrier in the work. If a student had an idea, Alex would encourage the student even if the topic did not concern Alex's own research. He had an encyclopaedic knowledge of the literature and, in the time before electronic databases, would provide bibliographical information when helpfully directing his students and others to the most relevant articles.

Some of Alex's former students and postdocs visited him regularly and maintained collaborations with him for decades, and Alex provided great personal, as well as professional, hospitality to them. Some of his visitors stayed multiple times in Alex's home for periods ranging from a few days to a couple of months. The Festschriften dedicated to Alex contain many paragraphs concerning his kindness and support and opportunities that he helped make possible. At the 2008 meeting in Alex's honour, one distinguished scientist told Hartquist of how, during his early career, he received even more support from Alex than from his own thesis supervisor. Alex must have spent a great deal of time writing letters of recommendation. He also expended considerable time visiting former students and postdocs, in part to support them in their new home institutions and, when invited to do so, to provide advice on how a department or research institute might develop its research programme.

Alex's help extended to teaching his students English composition. Hartquist will never forget Alex's written comment about 'dangling participles galore'.

The preceding comment gives an indication of some aspects of Alex's humour, which was incisive, dry, playfully interactive and always delivered in a friendly spirit.

Alex's own writing provides a model of concise, clear precision. He would sit quietly and quickly handwrite page after page with very few amendments. One did need some familiarity with his script to decipher it, but no editing was necessary.

Alex's sixtieth, seventieth and eightieth birthdays were celebrated with meetings held at Harvard, Cumberland Lodge in Windsor Great Park and Harvard, respectively. David Williams of UCL hosted the Cumberland Lodge meeting, while Alex's Harvard-Smithsonian colleagues and friends organized the other meetings. Some contributions to the four books and one special journal issue honouring Alex are cited in this memoir. The quantity of the meetings and Festschriften indicates the esteem in which Alex was held and reflects the affection that his colleagues and former students and postdocs had for him.

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AWARDS AND RECOGNITION

1969	Award of the International Academy of Quantum Molecular Science
1969	Fellow of the American Academy of Arts and Sciences
1977	Hodgkins Medal of the Smithsonian Institution
1980	Davison–Germer Award of the American Physical Society
1986	Gold Medal of the Royal Astronomical Society
1986	William F Meggers Award of the Optical Society of America
1989	Honorary Member of the Royal Irish Academy

1992	Spiers Memorial Medal of the Royal Society of Chemistry
1995	John A. Fleming Medal of the American Geophysical Union
1998	Asteroid 6941 named Asteroid Dalgarno with the approval of the International Astronomical Union
2001	Member of the National Academy of Sciences
2002	Hughes Medal of the Royal Society
2013	Benjamin Franklin Medal in Physics of the Franklin Institute

AUTHOR PROFILES

Thomas W. Hartquist

Tom Hartquist is Emeritus Professor of Astrophysics at the University of Leeds. Neal Lane, formerly a postdoctoral researcher for Alex in Belfast, supervised Tom's undergraduate research at Rice University. Tom met Alex in September 1974 when Tom arrived at Harvard. He began working with Alex in the summer of 1975, completed his PhD under Alex's supervision in January 1978 and worked as a postdoctoral researcher with Alex during two periods in 1978 and 1979. Alex encouraged Tom's interest in plasma and hydromagnetic processes, and they collaborated on inferring the interstellar cosmic-ray ionization rate from molecular observations and on shock chemistry and its effects on dynamics. Tom then spent four years at UCL, Alex's alma mater, before becoming an assistant professor of astronomy at the University of Maryland. From January 1985 to April 1998 he worked at the Max Planck Institut für Extraterrestrische Physik before moving to Leeds. Tom visited Alex once or twice each year until 2014 and often stayed in his home. They played tennis and squash together many times. Tom maintains his interest in molecular astrophysics and the dynamics of molecular regions. David Williams, a former Belfast student who worked with Alex, and Tom have collaborated closely.

Ewine F. van Dishoeck

Ewine van Dishoeck is Professor of Molecular Astrophysics at Leiden University, The Netherlands, where she studied chemistry and got her PhD in 1984. She gradually moved into astronomy during her PhD period under the co-supervision of Alex. Following postdoctoral and visiting positions at Harvard and Princeton, she was appointed as an assistant professor in cosmochemistry at the California Institute for Technology in 1988. In 1990, she moved back to Leiden University. The research of her group centres on the evolution of molecules from clouds to planet-forming discs, combining astronomy with studies of basic molecular gas-phase and grain-surface processes. She holds many international science policy functions, including president of the International Astronomical Union. She has been fortunate to receive prestigious awards, including the Dutch Spinoza award (2000) and the Kavli Prize for Astrophysics

(2018). She is a member of the Dutch, German and Norwegian Academies of Sciences, foreign associate of the US National Academy of Sciences, and foreign member of the UK Royal Astronomical Society.

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